LARGE LOW TEMPERATURE MAGNETORESISTANCE AND MAGNETIC ANOMALIES IN Tb_2PdSi_3 and Dy_2PdSi_3

R. Mallik, E.V. Sampathkumaran and P.L. Paulose
Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai-400005, INDIA.

The results of heat-capacity (C), magnetic susceptibility (χ), electrical resistivity (ρ) and magnetoresistance ($\Delta\rho/\rho$) measurements on the compounds, Tb_2PdSi_3 and Dy_2PdSi_3, are reported. The results establish that these compounds undergo long-range magnetic ordering (presumably with a complex magnetic structure) below (T_c=) 23 and 8 K respectively. The $\Delta\rho/\rho$ is negative in the vicinity of T_c and the magnitude grows as T_c is approached from higher temperature as in the case of well-known giant magnetoresistance systems (La manganite based perovskites); this is attributed to the formation of some kind of magnetic polarons. The magnitude of $\Delta\rho/\rho$ at low temperatures is quite large, for instance, about 30% in the presence of 60 kOe field at 5 K in Dy sample.

Among the ternary rare-earth (R) intermetallic compounds, only a few series of compounds of the type, R_2XY_3 (X= transition metals; Y=Si, Ga), crystallizing in AlB_2-derived hexagonal structures, has been known [1-8]. Recently, we have been paying special attention [8, 9] to the series, R_2PdSi_3 (Ref. 2). We reported the formation of Ce_2PdSi_3 (Ref. 8) and Eu_2PdSi_3 (Ref. 9), which were otherwise not known earlier. During the course of investigation of these Pd compounds, we have noted a minimum in the temperature dependent electrical resistivity (ρ) before long range magnetic ordering in the Gd alloy (Ref. 9), mimicking the behavior in magnetically ordering Kondo lattices, as a novel magnetic precursor effect. We have proposed that possibly the tendency for the localisation of conduction electrons due to exchange interaction (prior to magnetic ordering) is responsible for this behavior. From this result, we inferred that some kind of magnetic polaronic effects can be operative to some extent, not only in semiconductors [10, 11], but also in metallic systems [9] prior to long range magnetic ordering. In order to look for such effects in other compounds containing magnetic-moment carrying R ions and also as a continuation of our efforts to understand the magnetic behavior of this class of compounds, we have undertaken the investigations on Tb_2PdSi_3 and Dy_2PdSi_3.

The samples were prepared by arc melting stoichiometric amounts of constituent elements in an atmosphere of argon. The molten ingots were homogenised at 750 C for five days and the x-ray diffraction patterns confirm that these compounds crystallize in a AlB_2-derived hexagonal structure [2]. The heat-capacity (C) measurements were performed in the temperature interval 3 - 60 K by a semiadiabatic heat-pulse method [12]. The magnetic susceptibility (χ) data (2 - 300 K) were obtained employing a commercial superconducting quantum interference device in a magnetic field (H) of 2 kOe. Ac χ measurements (ac field 0.8 Oe) were also performed (4.2 - 100 K) at two different frequencies, 106 and 1090 Hz. The ρ data (2 - 300 K) were taken by a conventional four-probe method; in addition, the data were collected in the presence of a field of 50 kOe as a function of temperature (T up to about 100 K) and also as a function of H at selected temperatures (5, 10 and 35 K); the direction of the current is the same as that of H.

The results of C, χ and ρ measurements are shown for Tb_2PdSi_3 in Fig. 1. The C exhibits a distinct peak at 23 K, presumably arising from magnetic ordering. Above 25 K, there is an upward curvature of C and after subtraction of the lattice contribution employing the values of Y_2PdSi_3 (Ref. 8) following the procedure suggested by Blanco et al [13], we see a broad peak in the data (C_m), which is attributed to the Schottky anomalies arising from crystal-field effects. The low temperature χ data is plotted in the form of $d\chi/dT$ in Fig. 1b, which shows a minimum at 23 K and at the same temperature there is a drop in zero-field ρ (Fig. 1c). These results establish that Tb ions undergo long range magnetic ordering at 23 K (hereafter referred to as T_c). The plot of inverse χ versus T is linear above 30

K (Fig. 1d) and the effective moment ($\mu_{\rm eff}$) obtained from the slope of the plot turns out to be 9.87 μ_β typical of trivalent Tb ions. The Curie-Weiss parameter (θ_p) is found to be positive (11 K); while the positive sign indicates ferromagnetic ordering, the magnitude is lower than the actually observed value of T_c (= 23 K), as if antiferromagnetic correlations are also present. A careful look at the low temperature χ data (see inset, Fig. 1d) suggests that there is a tendency for χ to saturate below 10 K, possibly due to another magnetic transition. The plots (Fig. 1e) of isothermal magnetization (M) at 2 and 10 K are not linear, which might indicate ferromagnetic correlations; the saturation even at higher fields is absent; though the plots even above ordering temperature (30 and 35 K) look similar to those at 2 and 10 K presumably due to short range ferromagnetic correlations, the increase of M for initial application of the field is steeper at these low temperatures. The plots at 2 and 10 K look like a superposition of a saturated ferromagnetic component and a field dependent (antiferromagnetic) contribution. All these results indicate that the magnetic behavior of Tb_2PdSi_3 is quite complex and, in particular, the magnetic structure may not be of a simple ferromagnetic-type.

Now turning to the influence of the application of H on ρ, the ρ values are essentially unaltered by the presence of H above 60 K and hence we show the data only in the low temperature region. However, in the presence of a H (say, 50 kOe, Fig. 1c), the value of ρ gets gradually suppressed as T is decreased, rounding off the feature due to the magnetic ordering. This results in negative magnetoresistance [$\Delta\rho/\rho = \{\rho(H) - \rho(0)\}/\rho(0)$] in the vicinity of T_c, the magnitude of which increases gradually with decreasing T, attaining a large value of about -16% (for H= 50 kOe) at 20 K; we will return to the implications of this feature later in this article. We have also measured $\Delta\rho/\rho$ as a function of H at selected temperatures (see Fig. 1f); at 35 K, $\Delta\rho/\rho$ varies quadratically with initial application of H, which indicates the existence of spin-fluctuation effects; at 5 and 10 K, there is a sudden increase in the magnitude for small values of H, followed by flattenning in the range 10-20 kOe with a subsequent further increase in magnitude for H >30 kOe. This finding may be interpreted by the proposing that in zero field there is an antiferromagnetic component (also inferred from M versus H plots) undergoing metamagnetic transition with the increase of magnetic field. The negative sign of $\Delta\rho/\rho$ is consistent with ferromagnetic coupling.

With respect to the Dy sample, there is a clear-cut λ-anomaly below 8.5 K (Fig. 2a), arising from magnetic ordering from the Dy sub-lattice. The C_m, obtained as in the case of Tb sample, shows a broad peak presumably due to crystal-field effects. The plot of $d\chi/dT$ also exhibits a dip at 8 K (Fig. 2b) at the onset of magnetic ordering. The plot of inverse χ (Fig. 2d) is linear above 20 K and the value of μ_{eff} (10.5 μ_β) is typical of trivalent Dy ions. The value of θ_p is very close to zero and this indicates equal magnitudes of antiferromagnetic and ferromagnetic correlations. There is an upturn in χ below 7 K presumably due to another magnetic transition as in the case of Tb sample. The isothermal M (Fig. 2e) tends to saturate at 2 K, which implies dominance of ferromagnetism at high fields; however, the value of M at 55 kOe is far less than that expected for free Dy ion, which suggests that the magnetic ordering evolves from a crystalline electric-field-split level. With respect to ρ behavior, unlike in Tb sample, ρ shows an upturn (Fig. 2c) as the T is lowered across T_c; in the presence of H, however, the upturn vanishes; these findings establish [14] the formation of magnetic superzone boundary gaps, which suggests complex nature of the magnetic structure resulting in spin density wave formation. As in the case of Tb sample, ρ gets depressed continuously with decreasing T from few decades of T above T_c. It is interesting to note that the magnitude of $\Delta\rho/\rho$ is quite large at higher fields around T_c (see Fig. 2f), though far above T_c (say, at 35 K), it is close to zero.

We have also performed ac χ measurements at two different frequencies, 106 and 1090 Hz, on both these samples,

in order to look for possible spin-glass characteristics in these compounds. This becomes relevant considering that both antiferro- and ferro- magnetic correlations coexist in these alloys and that R ions form a triangular lattice [5]. The results are shown in Fig. 3. For these alloys, there is vitually no difference between the data recorded at two frequencies. For Tb sample, if at all there is a very small shift of the peak temperature with increasing frequency, it is towards low temperatures, in contrast to the expectation for spin-glass systems. Thus these results do not favor spin-glass freezing in the temperature range of investigation in these alloys. This is further confirmed by the occurence of the features due to magnetic ordering at exactly the same temperature in all the bulk measurements reported here (see the figures) for a given alloy which is not the case for spin-glass systems. We also note another interesting finding in the χ_{ac} data. There is an additional peak at about 15 K for the Tb case and at nearly the same temperature $d\chi/dT$ exhibits a shoulder (Fig. 1b); this finding is consistent with the proposal of another transition around 15 K. With respect to the Dy sample also, a broad peak at about 6 K below which χ tends to fall could be observed, though the transition at 8 K appears only as a shoulder due to proximity of these two transitions.

One of the points of main emphasis is that the magnetoresistance is negative in the vicinity of T_c, the magnitude of which increases as T is decreased from a few decades of T, in contrast to the general behavior of the influence of H on the conduction electrons to result in a small positive magnetoresistance. We have reported similar findings recently in some Gd compounds [15], which were interpreted in the following manner: there is a formation of polarised electron cloud around Gd ion due to strong s-f exchange interaction; the mobility of these electron clouds decreases with decreasing temperature possibly due to randomness of the magnetic exchange interaction, resulting from short-range magnetic order (and not in the sense of spin-glass freezing, which is not found in our alloys anyway); this immobile tendency contributes to electrical resistance as the long range magnetic ordering temperature is approached, in a way similar to the effect of crystallographic disorder resulting in weak electron localisation [16]. The application of external H aligns the randomly oriented spins (originated from short range magnetic order), which apparently facilitates free movement of carriers. This process of increase in conductance resulting in negative magnetoresistance is qualitatively similar to the mechanism generally proposed in the literature for giant magnetoresistance systems, viz., the ones based on La manganite perovskites, though double-exchange mechanism is not the one that mediates magnetic ordering in the rare-earth alloys. We call this as "magnetism-mediated electron localisation" (some kind of magnetic polaron, which has been invoked in it semiconducting rare-earth compounds, viz., EuSe [10], Gd pnictides [11]) and EuB_6 [17] as well as in GaAs-based semiconductors [18]). To our knowledge, the existence of weak electron localisation effects due to exchange interaction prior to long range magnetic order has not been recognised in *metallic systems*.

To conclude, in the compounds, Tb_2PdSi_3 and Dy_2PdSi_3, long range magnetic order sets in at 23 and 8 K respectively, presumably exhibiting complex magnetism. The χ data show features attributable to two magnetic transitions (the second one at about 15 and 7 K respectively) and it might arise from two crystallographically inequivalent sites [4]. It is of interest to perform neutron diffraction measurements to find out exact nature of the magnetic structures at these transition points. The weak electron-localisation effects attributable to a disorder in the exchange interaction seem to contribute to electrical resistivity prior to long range magnetic order, though the upturn in ρ is not apparent in the raw ρ data, resulting in a magnetoresistance behavior qualitatively similar to that noted for La manganite based perovskites. Thus, the magnitude of magnetoresistance is also quite large not only at lower temperatures, but also in the vicinity of magnetic ordering temperature (due to above factor) in our compounds. This work brings out the need to consider such magnetic precursor effects (electron locaisation mediated by disordered exchange interaction)

while interpreting the low temperature upturns in resistivity and heat-capacity in general even in *metallic* systems. This issue becomes relevant considering controversies in understanding the origin of the apparent non-Fermi liquid behavior in some f-electron systems [18].

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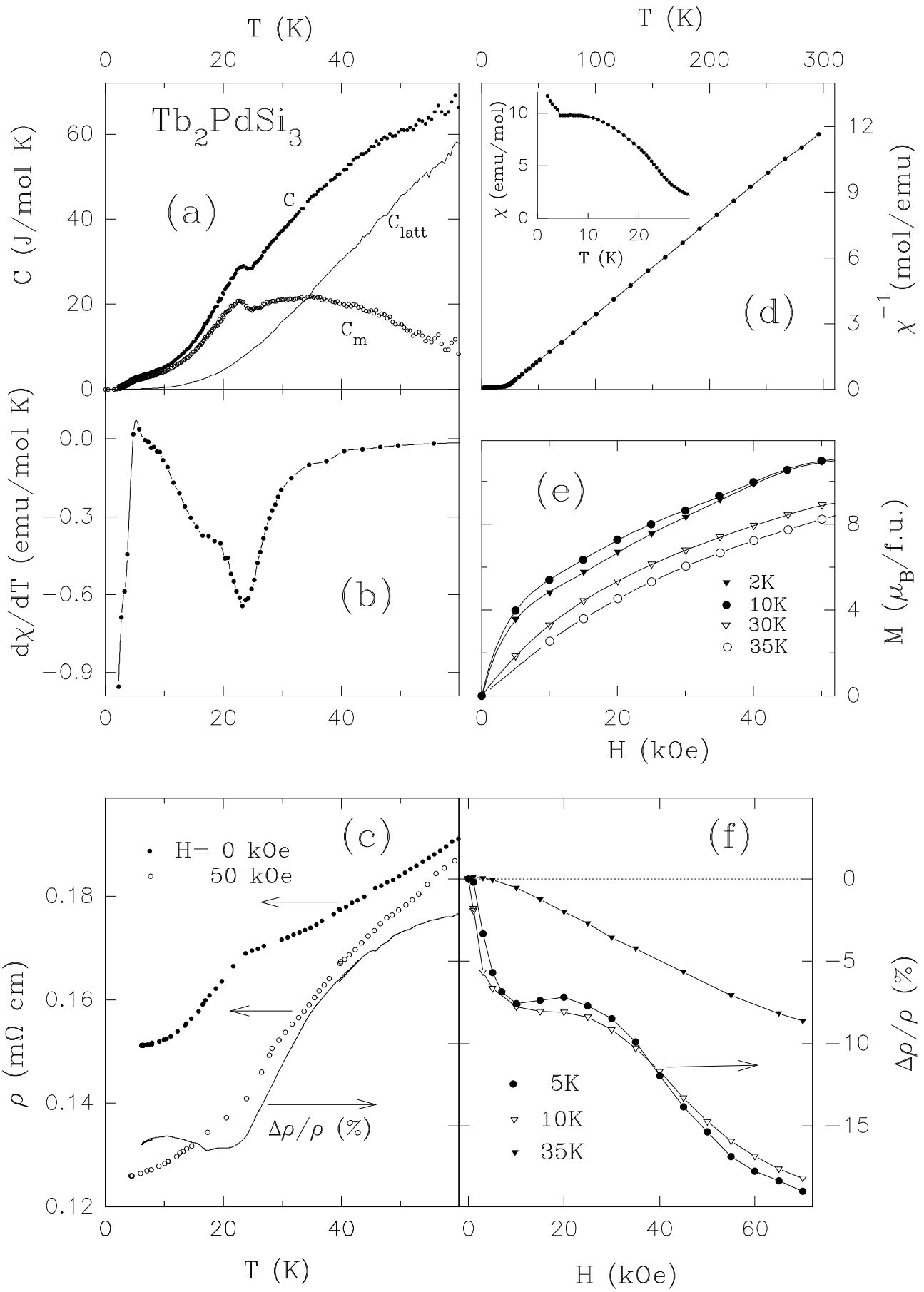
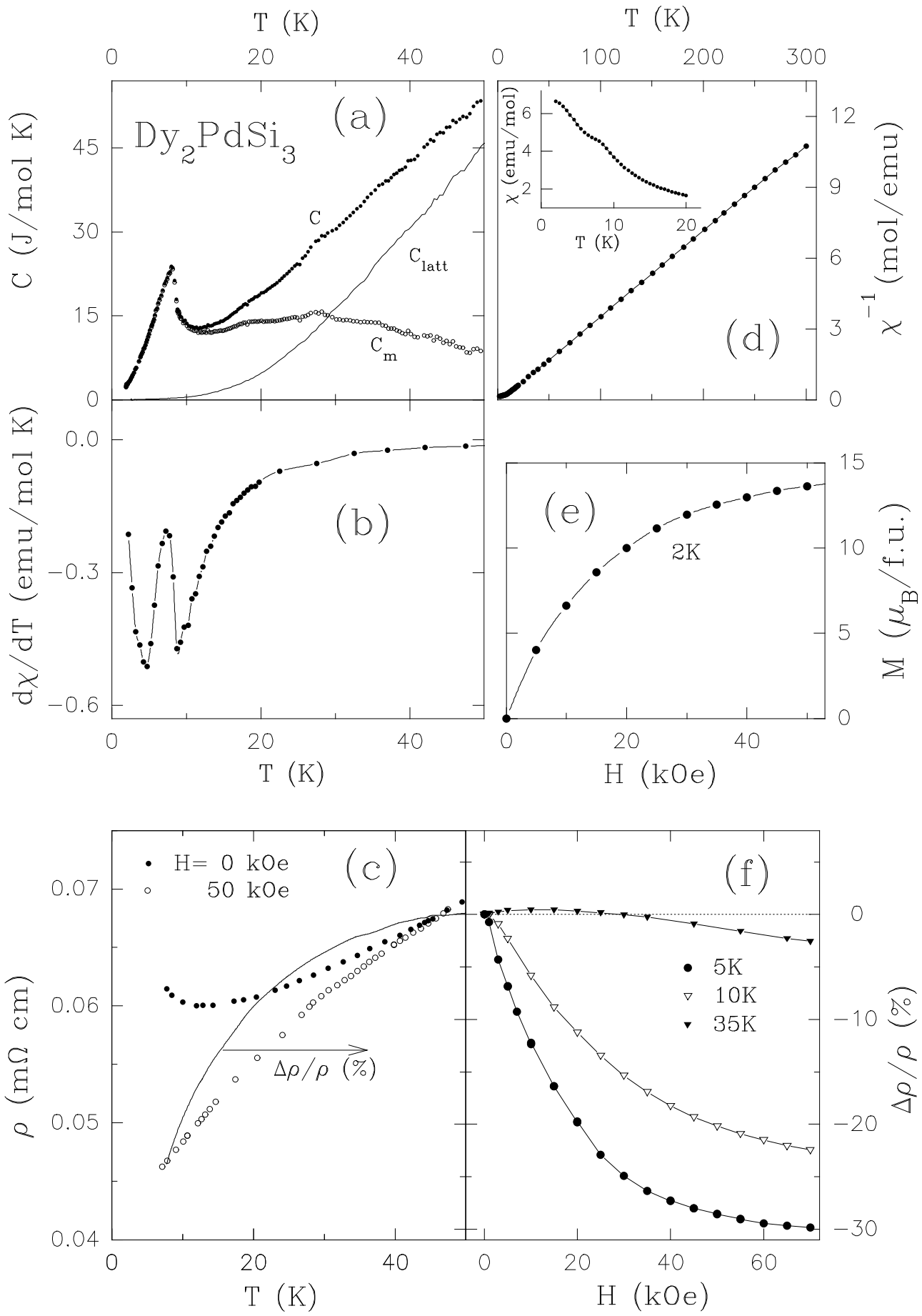
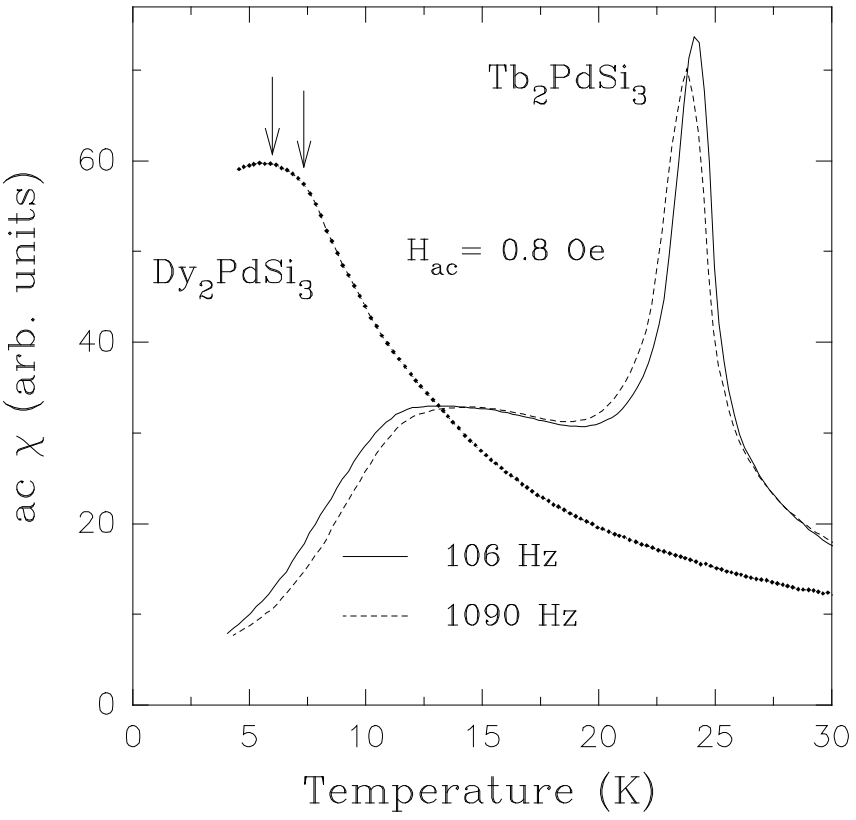
FIG. 1. (a) The heat-capacity, (b) the temperature derivative of magnetic susceptibility, (c) the electrical resistivity in zero field and in the presence of a magnetic field and magnetoresistance and (d) inverse susceptibility as a function of temperature for Tb_2PdSi_3. The isothermal magnetization and magnetoresistance as a function of magnetic field are shown in (e) and (f) respectively. In figure (a), the lattice and the magnetic contributions to C (see tect) in Tb_2PdSi_3 are also shown. Wherever the lines are drawn through the data points, these serve as guides to the eyes, except in (d), in which the line represents Curie-Weiss region. In the inset of Fig. (d), the low temperature χ data is plotted to highlight the features below 10 K.

FIG. 2. (a) The heat-capacity, (b) the temperature derivative of magnetic susceptibility, (c) the electrical resistivity in zero field and in the presence of a magnetic field and magnetoresistance and (d) inverse susceptibility as a function of temperature for Dy_2PdSi_3. The isothermal magnetization and magnetoresistance as a function of magnetic field are shown in (e) and (f) respectively. In figure (a), the lattice and the magnetic contributions to C (see text) in Dy_2PdSi_3 are also shown. Wherever the lines are drawn through the data points, these serve as guides to the eyes, except in (d), in which the line represents Curie-Weiss region. In the inset of Fig. (d), the low temperature χ data is plotted to highlight the features below 10 K.

FIG. 3. Ac susceptibility for Tb_2PdSi_3 and Dy_2PdSi_3 at two different frequencies in an ac field of 0.8 Oe. The approximate temperatures of two magnetic transition points for the Dy sample are marked by vertical arrows.


T (K)
Tb2PdSi3
(a)
C
C_latt
C_m
C (J/mol K)
(b)
dχ/dT (emu/mol K)
(c)
H= 0 kOe
50 kOe
Δρ/ρ (%)
ρ (mΩ cm)
T (K)
(d)
χ (emu/mol)
χ^-1 (mol/emu)
(e)
2K
10K
30K
35K
M (μB/f.u.)
H (kOe)
(f)
5K
10K
35K
Δρ/ρ (%)
H (kOe)


(a)
Dy_2PdSi_3
C
C_{latt}
C_m
C (J/mol K)
T (K)
(b)
dχ/dT (emu/mol K)
T (K)
(c)
H= 0 kOe
50 kOe
Δρ/ρ (%)
ρ (mΩ cm)
T (K)
(d)
χ (emu/mol)
T (K)
χ^{-1} (mol/emu)
(e)
2K
M (μ_B/f.u.)
H (kOe)
(f)
5K
10K
35K
Δρ/ρ (%)
H (kOe)


Tb2PdSi3
Dy2PdSi3
Hac= 0.8 Oe
106 Hz
1090 Hz
ac χ (arb. units)
Temperature (K)
0
20
40
60
0
5
10
15
20
25
30